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EVALUATION OF POLLUTION CONTROL IN FOSSIL FUEL CONVERSION PROCESSES. GASIFICATION: SECTION 5. BI-GAS PROCESS

C. E. Jahnig

Exxon Research and Engineering Company

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EVALUATION OF POLLUTION CONTROL IN FOSSIL FUEL CONVERSION PROCESSES

GASIFICATION: SECTION 5. BI-GAS PROCESS

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The report gives results of a review of the Bituminous Coal Research, Inc. BI-GAS Process, from the standpoint of its effect on the environment. The quantities of solid, liquid, and gaseous effluents were estimated, where possible, as well as the thermal efficiency of the process. For the purpose of reducing environmental impact, a number of possible process modifications or alternatives were proposed, and new technology needs pointed out.

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1. SUMMARY

The BI-GAS Process of Bituminous Coal Research, Inc. has been reviewed from the standpoint of its effect on the environment. The quantities of solid, liquid and gaseous effluents have been estimated, where possible, as well as the thermal efficiency of the process. For the purpose of reducing environmental impact, a number of possible process modifications or alternatives have been proposed and new technology needs have been pointed out.

TABLE OF CONVERSION UNITS

To Convert From	To	Multiply By
Btu	Calories, kg	0.25198
Btu/pound	Calories, kilogram	0.5552
Cubic feet/day	Cubic meters/day	0.028317
Feet	Meters	0.30480
Gallons/minute	Cubic meters/minute	0.0037854
Inches	Centimeters	2.5400
Pounds	Kilograms	0.45359
Pounds/Btu	Kilograms/calorie, kg	1.8001
Pounds/hour	Kilograms/hour	0.45359
Pounds/square inch	Kilograms/square centimeter	0.070307
Tons	Metric tons	0.90719
Tons/day	Metric tons/day	0.90719

2. INTRODUCTION

Along with improved control of air and water pollution, the country is faced with urgent needs for energy sources. To improve the energy situation, intensive efforts are under way to upgrade coal, the most plentiful domestic fuel, to liquid and gaseous fuels which give less pollution. Other processes are intended to convert liquid fuels to gas. A few of the coal gasification processes are already commercially proven, and several others are being developed in large pilot plants. These programs are extensive and will cost millions of dollars, but this is warranted by the projected high cost for commercial gasification plants and the wide application expected in order to meet national needs. Coal conversion is faced with potential pollution problems that are common to coal-burning electric utility power plants in addition to pollution problems peculiar to the conversion process. It is thus important to examine alternative conversion processes from the standpoint of pollution and thermal efficiencies, and these should be compared with direct coal utilization when applicable. This type of examination is needed well before plans are initiated for commercial applications. Therefore, the Environmental Protection Agency arranged for such a study to be made by Exxon Research & Engineering Company under Contract No. EPA-68-02-0629, using all available nonproprietary information.

The present study under the contract involves preliminary design work to assure that the processes are free from pollution where pollution abatement techniques are available, to determine the overall efficiency of the processes, and to point out areas where present technology and information are not available to assure that the processes are nonpolluting. This is one of a series of reports on different fuel conversion processes.

All significant input streams to the processes must be defined, as well as all effluents and their compositions. This requires complete mass and energy balances to define all gas, liquid, and solid streams. With this information, facilities for control of pollution can be examined and modified as required to meet Environmental Protection Agency objectives. Thermal efficiency is also calculated, since it indicates the amount of waste heat that must be rejected to ambient air and water and is related to the total pollution caused by the production of a given quantity of clean fuel.

Suggestions are included concerning technology gaps that exist for techniques to control pollution or conserve energy. Maximum use was made of the literature and information available from developers. Visits and/or contacts were made with the developers to update published information. Not included in this study are such areas as cost, economics, operability, etc. Coal mining and general offsite facilities are not within the scope of this study.

Considerable assistance was received in making this study, and we wish to acknowledge the help and information furnished by EPA, Bituminous Coal Research, Inc., and by Stearns-Roger Inc. The process design and balances used in this study are based on a detailed report on the BI-GAS process prepared by Air Products and Chemicals, Inc. (9).

Acknowledgement is also made to Dr. Henry Shaw of Exxon Research and Engineering who made the initial contacts to assemble background information on the BI-GAS process.

Four previous reports in this series on gasification were issued as Section 1 with the various process names listed. In reality they were Sections 1, 2, 3, and 4; thus, this report is labeled as Section 5.

3. BASIS AND BACKGROUND

A number of processes have been evaluated for making clean fuel from coal (1,2,3,4,5,6). These include gasification at pressures from near atmospheric in the case of Koppers-Totzek, to 1,000 psig, for example with BI-GAS or Synthane. Reaction temperatures also cover a range of from moderate temperatures in the Lurgi process, to very high temperatures in the Koppers-Totzek process with slagging of the ash. Some processes such as Lurgi and Synthane make by-product char or tar, while others make no by-products (CO_2 Acceptor and Koppers-Totzek processes). The BI-GAS process avoids making by-product char or tar, using a two zone gasifier with the upper zone at 1700°F, while the lower zone is at 3000°F and produces slag from the coal ash.

As a result of early studies, Bituminous Coal Research concluded that an optimum type gasification process would have the following features:

(1) Operation at high pressure to avoid the need for compression when supplying pipeline gas.

(2) Make no char by-product.

(3) No tars or liquid products would be produced which would complicate the clean-up.

This led to the concept of a high-pressure, two zone gasifier. Temperature of the upper zone is high enough to prevent tar formation, while the lower zone is at 3000°F so that residual slag is low in carbon content and can be discarded. A further advantage of high pressure is that it increases the amount of methane formed in the gasifier, thereby significantly reducing the heat load on the gasifier and the gas volume to be handled in the downstream operations.

Information is available in the literature on the BI-GAS process, including the design of the pilot plant facilities (7,8), and projections of a commercial plant design and operation (9). In the present study to evaluate environmental aspects of the process, we have used as a starting basis the commercial plant projections developed by Air Products and Chemicals, Inc. (9). Some modifications were made where necessary to assure environmentally sound operations, as for example, to reduce sulfur emissions from coal fired furnaces. Also, in the course of the study, other modifications became apparent which could give better environmental control or improve thermal efficiency of the process, and these are described briefly in this report for consideration.

The plant is sized to make 250 million SCFD of pipeline gas by gasifying coal with steam and oxygen. The design includes shift conversion and methanation to give a gas with a heating value of 943 Btu per cubic foot, available at 1,075 psia. Western Kentucky coal is used, and after cleaning and washing, the amount is 14,535 tons per day (at a nominal 8.4% moisture) which provides all of the fuel for coal drying and utilities production in addition to the gasification requirements.

4. PROCESS DESCRIPTION

A flow plan of the process is shown in Figure 1, together with major flow rates and operating conditions. Coal used is shown in Table 1, while products are shown in Table 2. It is convenient to subdivide the process into the following operations, each of which will be described in the following subsections: (1) Coal Preparation, (2) Gasification, (3) Quench and Dust Removal, (4) Shift Conversion, (5) Acid Gas Removal, (6) Methanation, and (7) Auxiliary Facilities.

4.1 Coal Preparation and Drying

This process section includes crushing, cleaning and drying as well as a storage pile with 30 days capacity. Run of mine coal feed amounts to 23,243 tons per day. This is crushed and coarse refuse is rejected amounting to 4,804 tons per day. The coal can then be sent to storage, or to the washing operation which rejects an additional 3,904 tons per day. Drained coal from washing, containing 8.4% moisture, is used partly as fuel to the utilities plant supplying steam for the process, while the remainder goes to the grinding and drying facilities. Here it is ground to 70% smaller than 200 mesh, dried to 1.3% moisture, and sent to storage silos. Some of the dried coal is used as fuel in the dryer, amounting to 11,137 pounds per hour or about 134 tons per day.

Since the gasifier operates at 80 atmospheres, it is necessary to pressurize the coal feed. The original design used piston feeders to push the coal into a high pressure feed hopper and is the system used in the present environmental evaluation. Subsequent work has indicated that other methods such as lock hoppers or slurry feeding may be preferable; however. the change would make only minor modifications in effluents to the environment, although thermal efficiency would be lower than for the case using piston feeders.

4.2 Gasification

The coal is gasified using steam and oxygen in a two zone reactor at 80 atmospheres. Operation of the reactor is based on entrained flow rather than using a fluidized bed or fixed bed reactor. Coal is fed to the top 1700 F zone where it mixes with steam and hot synthesis gas entering from the lower zone. Conditions in this upper zone favor high formation of methane, with negligible amounts of tar or oil. Although the volatile content of the coal feed is completely consumed, there is considerable unreacted char remaining which is carried out with the gas and recovered by cyclones following the reactor.

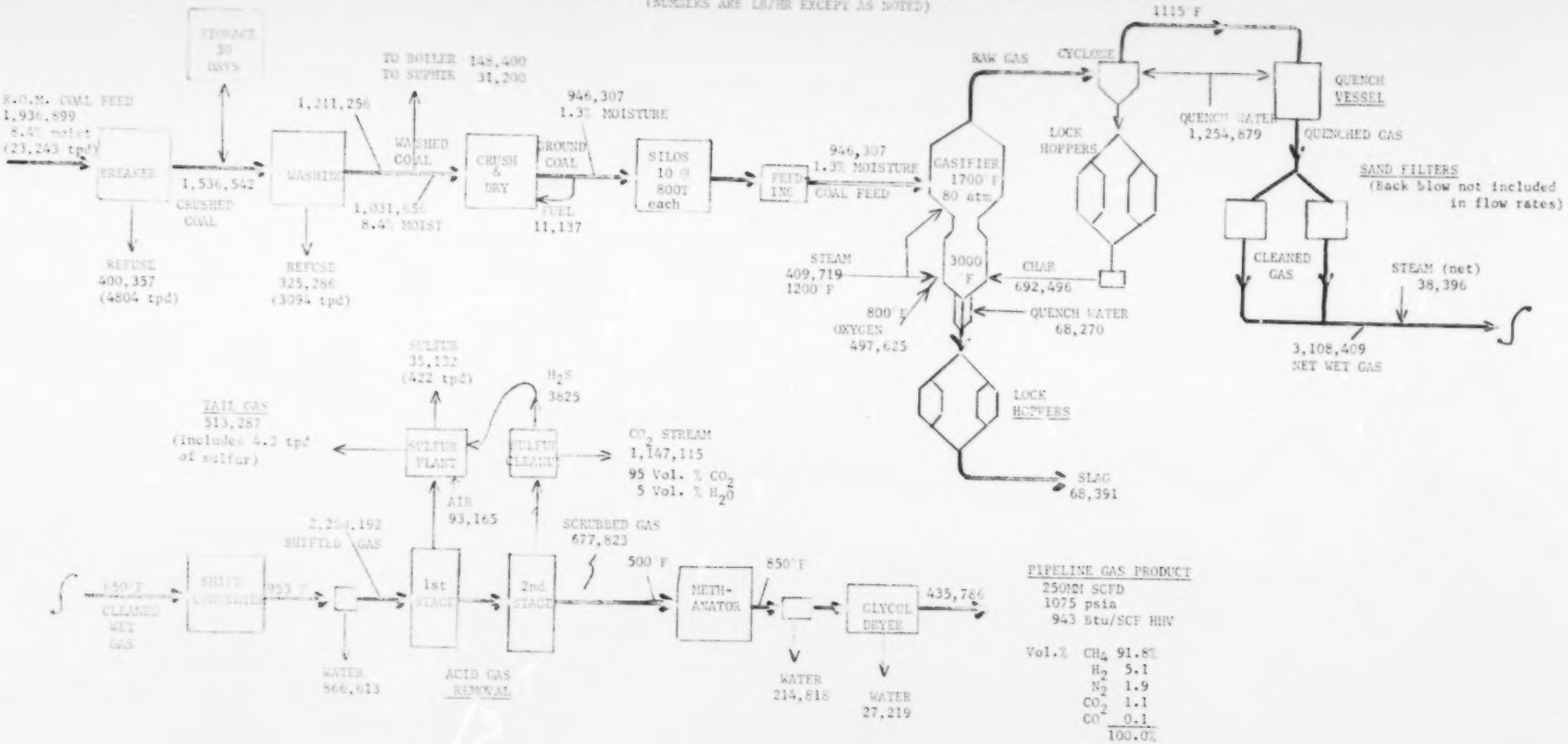
The char is recycled by means of lock hoppers to the lower gasification zone where it is reacted with steam and oxygen at 3000 F. A special char feeding system is provided, since it is indicated that a reliable and very uniform feed rate must be maintained, so as to avoid conditions that could give excessive flame temperatures. Synthesis gas is formed and passes to the upper reactor as described earlier. Slag is withdrawn from the bottom, quenched with water, and removed by way of lock hoppers. Since it has little or no combustible content, it can be discarded (from an energy viewpoint).

Figure 1

BIGAS - PROCESS

FLOWPLAN AND FLOW RATES FOR PLANT MAKING 250MM SCFD OF PIPELINE GAS FROM W. KENTUCKY NO. 11 COAL

(NUMBERS ARE LB/HR EXCEPT AS NOTED)



PIPELINE GAS PRODUCT
250MM SCFD
1075 psia
943 Btu/SCF HHV

Vol.%	CH_4	91.8%
	H_2	5.1
	N_2	1.9
	CO_2	1.1
	CO	0.1
		100.0%

Table 1

Coal Feed - W. Kentucky No. 11

	As Rec'd.	Dried	Moist & Ash Free
Proximate Analysis Wt %			
Moisture	8.4	1.3	--
Volatile matter	39.5	42.5	46.5
Fixed carbon	45.4	49.0	53.5
Ash	6.7	7.2	--
Ultimate Analysis Wt. %			
Carbon	68.15	73.40	80.20
Hydrogen	4.67	5.03	5.50
Nitrogen	1.37	1.48	1.62
Sulfur	3.48	3.75	4.10
Oxygen (by diff.)	7.24	7.84	8.58
	84.91	91.50	100.00
Heating Value			
HHV Btu/lb.	12,330	13,285	14,510

Coal Consumed	% Moisture	lb/hr
Coal Dryer	1.3	11,137
Gasification	1.3	946,307
Utility Boiler	8.4	148,400
Steam Superheater	8.4	31,200

Table 2

Products from BI-GAS Process

Pipeline gas	
Volume, MM SCFD	250
Pressure psia	1075
Temperature °F	95
High Heating Value Btu/SCF	943
Composition vol%	
CH_4	91.8
H_2	5.1
N_2	1.9
CO_2	1.1
CO	0.1
	100.0
By product Sulfur tpd	422
Slag (dry basis) tpd	
Gasifier	820
Dryer	10
Boiler + Superheater	144
	974
NH_3 potential tpd	112
(@ 60% of N in coal)	

4.3 Quench and Dust Removal

Hot raw gas from the gasifier passes to cyclone separators which remove most of the char and solid particles in the gas. Quench water is added to the cyclone in order to moderate the temperature, and additional quench water is added in a quench vessel after the cyclone separator.

The quenched gas still contains some dust that was not removed by the cyclones, but must be removed so as not to plug the fixed bed of shift conversion catalyst. Rather than scrub the dust out with water, which would require considerable cooling, the dust is filtered out at high temperature using sand beds. These operate in parallel in a cyclic manner. Pressure drop will build-up during the onstream cycle, and the bed is cleaned when necessary by back flushing with clean gas so as to lift and agitate the sand particles. Entrained dust from back flushing is then returned to the gasifier where it leaves with the slag.

4.4 Shift Conversion

After dust removal, the gas next goes to a shift converter where carbon monoxide reacts with steam to form hydrogen and carbon dioxide, increasing the ratio of H_2 to CO to three to one as required in the final methanation. A sulfur resistant shift catalyst must be used, resulting in relatively low activity compared to those used on sulfur free gases. A large excess of steam is maintained to give 50 mol. % steam in order to facilitate the desired reaction and to prevent catalyst degradation or carbonaceous deposits. Steam conversion in this shift reactor is about 27%.

After shift conversion, the gas is cooled to remove most of the remaining moisture. This, of course, produces sour water containing H_2S and ammonia and possibly traces of cyanides, phenols, etc. It is conveniently disposed of by using it as part of the quench water, and thereby provides steam required for shift conversion. One advantage of this specific design is that a very large quantity of sour water can be disposed of by injecting it into the hot gas for quenching. A further advantage is that no facilities are then needed for generating steam used in shift conversion, and neither are exchangers needed for cooling the hot raw gas from the gasifier.

4.5. Acid Gas Removal

Removal of all sulfur compounds is needed to meet pipeline gas specifications and to protect the methanation catalyst. The bulk of the sulfur, as well as CO_2, is removed using the proprietary Benfield process based on hot carbonate scrubbing. Two separate absorber towers are used in series. The first of these produces a gas relatively high in sulfur content, about 8% H_2S, to facilitate sulfur recovery in the Claus plant. The second absorber is for final cleanup of sulfur from the gas and for CO_2 removal.

Most of the CO_2 is removed in this second absorber and vented to the air; however, this CO_2 vent stream contains excessive amounts of H_2S, namely 3400 ppm, and further processing is needed to clean it up. Therefore, adsorption using molecular sieves has been provided to recover the H_2S content and send it to the Claus sulfur plant. Air Products has indicated (9) that a Rectisol process which uses scrubbing with refrigerated methanol would be preferable for acid gas removal, and that it would produce a reasonably concentrated stream of H_2S for the Claus plant while at the same time giving a clean CO_2 stream which could be vented directly to the air. However, other studies indicate that this vent stream from a Rectisol unit requires incineration or cleanup because of excessive content of combustibles and sulfur (5). The Rectisol process uses methanol scrubbing at low temperature, and can remove carbonyl sulfide and other contaminants. Gas leaving the hot carbonate scrubbing system used in the present design contains moisture, most of which is removed by cooling the gas ahead of methanation. This is a clean condensate which can be used for boiler feed water make-up.

Gasification can produce many compounds in addition to H_2S, such as cyanides and thiocyanates as well as large amounts of ammonia. There are also various sulfur compounds, particularly carbonyl sulfide and some carbon disulfide. It is essential to completely remove all of these before methanation in order to protect catatlyst activity. Most of the ammonia and compounds that are highly soluble in water will be removed in the condensation after shift conversion. Hot carbonate systems for acid gas removal have the important advantage that they do remove carbonyl sulfide. Amine systems, in general, do not remove carbonyl sulfide, and moreover react irreversibly with cyanides thus requiring purge of the chemical solution.

4.6 Methanation and Drying

Clean synthesis gas is methanated in this section to increase the heating value of the gas up to pipeline quality. The reaction of CO with 3 volumes of H_2 to make methane and water can be carried out in a fixed bed of nickel catalyst. A guard bed of zinc oxide ahead of the reactor removes traces of sulfur compounds in order to protect the methanation catalyst. Methanation is a highly exothermic reaction, releasing about 20% of the heating value in the reacting gases. Reactor temperatures of 500°F at the inlet and 850°F at the outlet are maintained by recirculating some of the gas leaving the reactor through exchangers to generate high pressure steam. Methanation is carried out to a high conversion so that the residual CO content is no more than the 0.1 Vol. % specified for pipeline quality gas. Residual hydrogen content is 5.1 Vol. %. Since methanation generates a considerable amount of water, this is recovered as clean condensate upon cooling. More complete drying of the gas is then carried out using a glycol system to meet the requirement of 7 lb water maximum per MM SCF of gas.

For the present study the processing sequence used by Air Products has been followed. Their flow rates and utility requirements were reviewed and used in the evaluation of environmental aspects.

4.7 Auxiliary Facilities

In addition to the gasification system, auxiliary facilities are needed to make the plant complete and self-sufficient. A Claus plant is included to make by-product sulfur from the H_2S that is recovered in acid gas removal. The basic Claus plant will not give adequate sulfur recovery or clean-up, since the feed gas will contain no more than 15% H_2S, therefore tail gas clean-up was added.

A conventional air separation plant is included in the base design to provide oxygen needed for gasification. It does not generate contaminated waste streams, but it is a large consumer of utilities and therefore has an important effect on thermal efficiency.

As would be expected, the process uses large amounts of steam and electricity. All utilities needed to make the plant self-sufficient are provided in the design, including high pressure and low pressure steam, electric power generation, water make-up treating, circulating cooling water, and waste water treating. Fuel requirement for these has been been included on the basis that coal would be used for fuel. Since the coal has a high sulfur content, pollution control will be needed on these fuel consumers. The simplest approach is to add flue gas clean-up so that coal can still be used as fuel, and a number of processes are available (10). An alternative would be to use low sulfur, low Btu gas made in the process for fuel in utilities generation and in coal drying.

The particular study includes utilities requirements for offices, shops, laboratories, and cafeteria (e.g. 50,000 lb/hr of steam for heating buildings). These are not always included in similar studies of other processes; therefore, caution is required in making comparisons with other studies.

5. EFFLUENTS TO AIR

Overall flow rates for the process were shown in Figure 1. Figure 2 and Table 3 show all of the streams entering and leaving specific units, some of which are returned to other units within the plant. All streams which are actually discharged to the environment are indicated by heavy dashed lines in Figure 2 and by asterisks in Table 3. For discussion these are grouped according to whether they are released to the air or represent liquid and solid effluents. Effluents to the air are discussed in the following subsections.

5.1 Coal Preparation and Drying

The first effluent to the air is from the coal handling and preparation area. Run of mine coal is delivered by rail and truck and conveyed to a breaker where it is crushed to 1-1/2 inches and smaller. Refuse amounting to 4804 tpd is rejected at this point and must be disposed of in a suitable manner. Such operations will normally have a dust problem, and careful consideration and planning is required for control. Covered conveyers should be provided wherever possible; even so, there may be vent streams or leaks that could release dust. If needed, a dust collection system could be used operating at slightly below atmospheric pressure to collect vent gas and pass it through bag filters. Since spills from conveyers and leaks can also create dust, facilities such as clean-up equipment and water sprays may be needed.

The coal storage pile is also of concern in that wind can pick up and disperse fine particles. Evaluation is needed for each specific situation in order to provide proper control measures. Proposals for dust control have been made such as spraying oil or asphalt on the surface of the pile, or covering it with plastic. The amount of coal handled is so large that a loss of even a small fraction of a percent could be excessive.

A further consideration on any coal storage pile is the possibility of fires and spontaneous combustion which would result in evolution of odors, fumes, and volatiles. One control measure is to compact the pile by layers as it is being formed. In any event, plans and facilities should be available for extinguishing fires if they occur (11).

The next step is to wash and screen the coal, and in this operation another 3904 tpd of refuse is rejected. Disposal of this refuse should be carried out in a way to avoid pollution. Since it is wet there should be little or no dusting problem except when it dries out. However, it can be expected that there will be spills of the refuse or coal in the coal preparation area, and that these will create a dust nuisance when they dry out and are disturbed by the wind or by trucks. Again this calls for plans and facilities for cleaning up dust and for flushing to the storm sewers. Although a detailed design of this coal preparation and handling system is not available, it will no doubt include a tailing pond to allow recovery and disposal of fine material from the washing operation. Proper environmental controls are needed as discussed in the literature (12).

Figure 2
BIGAS PROCESS

BLOCK DIAGRAM SHOWING STEAMS IN & OUT OF SPECIFIC SECTIONS OF PLANT

(Emissions to environment are indicated by heavy dashed line, other streams are returned to process)

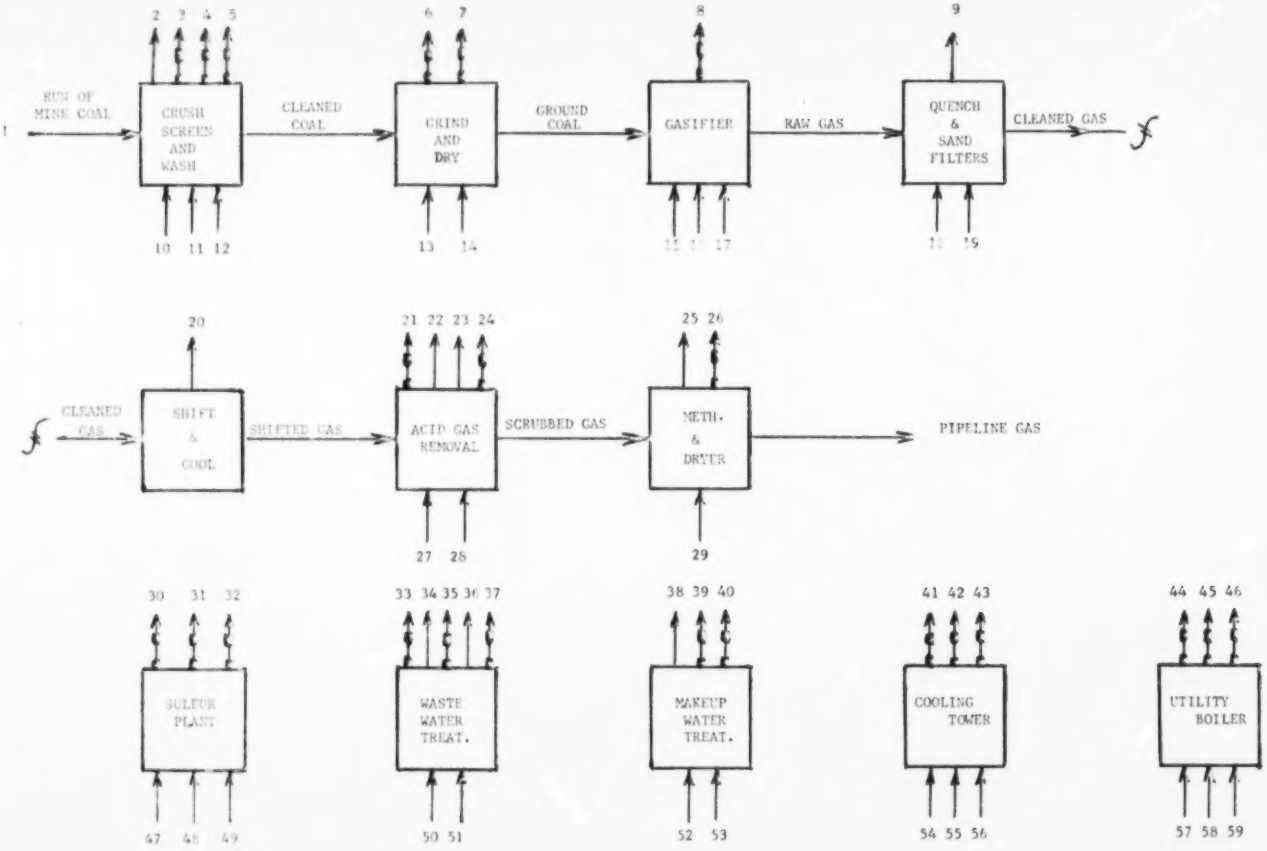


Table 3

BI-GAS Process - Inputs and Effluents

Stream Number	Identification	Flow Rate lb/hr	Comments
1	Coal to gasifier	946,307	Dried cleaned coal 1.3% moisture
2	Rain runoff	e.g. 6" in 24 hrs.	Runoff from coal storage pile
3 *	Wind	--	Can cause dust nuisance
4 *	Refuse	400,357	Rock waste from coal cleaning
5 *	Wash water	e.g. 4000 gpm	Recirculate through clarifiers and tailing pond for cleanup & reuse.
6 *	Flue gas	131,700	Vent gas from coal dryer. (For analyses see Table 4)
7 *	Spent limestone	2,970	From vent gas cleanup on coal dryer, e.g. spent limestone plus ash from coal fuel
8 *	Slag from gasifier	68,391	Plus equal wt. of water to form slurry for handling
9	Dust recovered by sand filters	--	Returned to gasifier by back blowing with part of cleaned gas, e.g. 5% of total gas
10	Rain	e.g. 6" in 24 hrs.	Rain on coal storage and handling area
11	Wind	--	Wind effect on coal storage and handling area
12	Wash water	e.g. 4000 gpm	Used to clean crushed coal
13	Air	122,700	For burner on coal dryer
14	Limestone	2,500	Raw materials used for stack gas cleanup
15	Steam	409,719	To gasifier to react with coal
16	Oxygen	497,625	Oxygen to gasifier to generate heat
17	Quench water	68,270	To quench 3000°F slag--steam is returned to gasifier
18	Sand	--	Makeup to sand bed filter
19	Quench water	1,254,879	Quenches 1700°F raw gas from gasifier, includes sour water

Table 3 (Cont.)

BI-GAS Process - Inputs and Effluents

Stream Number	Identification	Flow Rate lb/hr	Comments
20	Sour water	866,613	Condensed from raw gas, contains H_2S, NH_3 etc. Is returned to Quench (#19 above)
21*	Chemical purge	e.g. 50,000	Purged to reject contaminants from acid gas scrubbing solution. Will contain potassium carbonate
22	H_2S stream	451,429	Acid gas sent to Claus plant for sulfur recovery (6.9 vol. % H_2S, 44.7 vol. % CO_2, 48.4 vol. % H_2O)
23	H_2S (pure)	3,825	To Claus plant. From molecular sieve recovery on CO_2 vent gas.
24*	CO_2 vent stream	1,147,115	From acid gas removal (mol. sieve is used to control sulfur emission).
25	Water	214,818	Formed by methanation reaction
26*	Water reject	27,219	Removed by glycol dryer on product gas
27	Chemical makeup	--	Makeup chemicals to acid gas remove system. Will include K_2CO_3 and possibly inhibitors, antifoam agents, etc.
28	Molecular sieve	--	Makeup on sieve to clean up CO_2 vent stream
29	Glycol	--	Makeup agents to glycol dryer
30*	Tail gas	513,287	From sulfur plant after tail gas cleanup
31*	Sulfur	35,132	By-product sulfur recovered
32*	Purge chem.	--	From tail gas cleanup operation, purged to reject contaminants
33*	Ammonia	7,689	Potential by-product separated in waste water treating
34	H_2S	e.g. 87	Stripped out of sour water and sent to sulfur plant.
35*	Phenols	--	Potential by-product or to disposal from waste water treating -- amount unknown

Table 3 (Cont.)

BI-GAS Process - Inputs and Effluents

Stream Number	Identification	Flow Rate lb/hr	Comments
36	Treated water	86,000	Water after treating. A small stream of sour water will probably have to be purged to reject contaminants and trace elements from the system. Further information is needed to define cleanup requirements.
37*	Sludge	see Table 13	Sludge formed in waste water treating, e.g. from biox, misc. solids. May have odor problem. Should be incinerated.
38	Makeup water	3,489,000	To cooling tower and boiler feed water
39*	Chemical waste	see Table 13	Chemicals used in treating makeup water
40*	Sludge	see Table 13	Sludge formed in treating water makeup with lime, alum., etc. can be disposed of with slag.
41*	Air	272,000,000 (85 MMM SCFD)	Air flowing through cooling tower (plus evaporated water 2,626,000 lb/hr)
42*	Water mist	e.g. 263,000	Drift loss from cooling tower (0.2% of circl.)
43*	Water	ca. 600,000	Blowdown from cooling tower
44*	Flue gas	1,971,000	From utility boiler (see Table 4).
45*	Slag	12,033	From coal used as fuel on utility boiler (may dispose of with gasifier slag)
46*	Spent limestone	44,530	Used to desulfurize flue gas on utility furnaces
47	H_2S stream	455,254	To sulfur plant for recovery (Streams 22 and 23)
48	Air	93,165	Used in Claus plant to burn H_2S
49	Chemical	--	To tail gas cleanup on Claus plant.
50	Sour water	e.g. 86,000	Purge of sour water may be required to prevent build up of trace elements etc. in recirculated sour water.
51	Chemicals	see Table 13	As required to clean up purge stream of sour water.

Table 3 (Cont.)

BI-GAS Process - Inputs and Effluents

Stream Number	Identification	Flow Rate lb/hr	Comments
52	Makeup water	3,489,000	Treated and used as makeup for cooling tower and boiler feed water
53	Chemicals	see Table 13	Used to treat makeup water, e.g. lime, alum, caustic, acid, ion exchange resin
54	Air	272,000,000 (85 MMM SCFD)	Air flow through cooling tower
55	Cooling water	262,580 gpm	Recirculated cooling water
56	Chemicals	see Table 13	Antifouling (e.g. chlorine) and anticorrosion (e.g. chromate) agents in cooling water circuit.
57	Air	1,837,000	Combustion air used in utility furnaces
58	Coal	179,600	Coal fuel used in utility boiler pl superheater (8.4% moisture)
59	Limestone	37,500	Used for stack gas cleanup on utility furnaces

* Streams emitted to the environment.
Other streams are returned to process.

Noise control should be carefully considered since it is often a serious problem in solids handling and size reduction. If the grinding equipment is within a building, the process area may be shielded from undue noise, but additional precautions are needed for personnel inside the building.

Following the washing operation, cleaned coal is sent to the crushing and drying system and also to the utility areas as fuel. Adequate dust control is needed on all these handling operations. In the dryer, moisture content is reduced from 8.4% to 1.3% by contacting with hot flue gas. Heat is supplied by burning part of the dried coal. Since this fuel has 3.75% sulfur, corresponding to 5.64 lb SO_2 per MM Btu, it will be necessary to clean up the vent gas to remove sulfur as well as particulates. It will be desirable to recover and use the coal fines, for example, by using dry cyclones, then sulfur in the flue gas can be removed by one of the processes that are offered for stack gas clean-up (13). Some of these have proposed to use a throw-a-way limestone medium, while others provide for regeneration of a chemical scrubbing agent to make by-product sulfur, sulfuric acid, or gypsum.

In the drying operation a large volume of hot gas is contacted with the coal. Oxygen content is normally limited to about 10 Vol. % by safety considerations. Also the maximum temperature should be limited to avoid heating the coal above 500°F, so as not to release volatile matter. It is common practice to use a large amount of excess air, such as 100%, in order to minimize moisture content of the drying gas and thereby facilitate drying. In some cases effluent gas may be recycled or inert gas added to control gas temperature and oxygen content.

With the present high price of fuel, the design of drying facilities should be reconsidered and optimized to minimize fuel consumption. This subject is discussed more fully in a previous study (4). In brief, it is desirable to operate the dryer with minimum excess air, for example 10% excess, and to recycle vent gas as needed to control temperature of the hot gas. This gives minimum fuel consumption as well as minimum volume of vent gas to be cleaned up. Of course, the moisture content of the drying gas will be higher than when a large amount of excess air is used making it more difficult to achieve the same degree of drying, although the moisture content of the dried coal could be allowed to increase slightly. Further details on flue gas composition are given in Table 4 and accompanying notes.

In general, it will be desirable to maximize the preheat temperature on the coal feed, and to preserve this sensible heat so as to reduce heat load on the gasifier and reduce oxygen requirement. Preheat temperatures as high as 500°F have been used without substantial evolution of volatile matter from coal. This temperature has also been considered practical from the standpoint of using lock hoppers.

The coal feeding system for pressurizing the coal in this specific design is based on a piston feeder as originally proposed. Storage silos are also included. Normally there will be no effluent to the air from this system, although it may involve pneumatic transport of coal, in which case

Table 4

Flue Gas Flow Rates and Composition
From Boiler Plus Steam Superheater

Fuel Fired (Alternatives)	Coal	Low Btu Gas
Fuel lb/hr	179,600 (8.4% Moist)	132,663
Air lb/hr	1,837,000	1,538,000
Flue Gas lb/hr	1,971,000 (586 MM SCFD)	1,725,000 (562 MM SCFD)
Flue Gas Comp. Vol. %		
CO_2	15.5	8.3
H_2O	6.4	21.0
SO_2	0.3	--
N_2	76.0	69.0
O_2	1.8	1.7
	100.0	100.0

NOTES:

(1) Sulfur contained in above coal amounts to 6,250 lb/hr, and flue gas cleanup must be provided. Using limestone scrubbing, for example, would require 37,500 lb/hr of limestone, at twice the theoretical consumption.

(2) On coal dryer, flue gas composition from combustion of coal fuel will be similar to above. In addition, moisture amounting to 74,212 lb/hr is removed from coal, giving a total of 51% H_2O in vent gas.

(3) Ash from coal used for fuel to be disposed of:

Coal dryer	803 lb/hr
Boiler plus Superheater	12,033 lb/hr

(4) High heating values are: 12,330 Btu/lb for coal, and 16,695 Btu/lb for low Btu gas.

recovery and clean-up of the conveying gas is needed. In the event that lock hoppers are used instead of the piston feeder, then there will be considerably more vent gas from depressuring the hoppers which should be cleaned-up and returned to the system. A promising way to reduce the volume of gas from the lock hopper operation is to use pressure stages so that the highest pressure gas from the final stage can be used to pressure the initial lock hopper stage to an intermediate pressure level.

Instead of using stack gas clean-up on the coal dryer for sulfur control, it would be possible to use part of the low Btu product gas for fuel. Dust removal on the vent gas could then be by bag filters or a scrubber. This route would, of course, call for an increased capacity on the gasification system.

5.2 Gasification

In normal operation there will be no effluents to the air from the gasification section, since all of the gas streams are contained and processed in downstream equipment.

Slag formed in the lower zone of the gasifier is quenched with water and the resulting steam flows back up into the gasifier. Quenched slag is removed by way of lock hoppers. It is handled as a water slurry, and reliance is placed in the shattering effect of the quench to control particle size of the slag and provide a slurry that can be handled. Typically, the slurry may contain equal weights of slag and water. Depending on the final disposition of the slag slurry, there may or may not be a dust problem. For example, if it is used as land fill or if it goes to a storage pile, there could be a dust problem when it dries out. The possibility of odors needs to be defined for the handling and disposal system. Also, other emissions will occur and need to be defined.

5.3 Quench and Dust Removal

Raw gas leaving the gasifier goes through cyclones to recover entrained dust or char, which is then returned to the lower stage of the gasifier by means of lock hoppers. It should be possible to contain this system and the pressurizing gas so that normally there will be no emissions to the air.

After quenching, the gas goes through sand bed filters in parallel to remove dust. These filters are cleaned by back blowing with part of the synthesis gas, and this dusty stream is returned to the gasifier for disposal and thereby contained within the system.

Normal maintenance will be needed on the sand filters and possibly also as a result of upsets. Precautions are needed to control emissions to the air during such periods, for example, in cleaning or replacing the sand and on depressuring the equipment. Gas released on depressuring should be recovered and returned to the system. Similar comments apply to the lock hoppers and other parts of the process with regard to depressuring and maintenance.

5.4 Shift Conversion

Shift conversion does not generate gaseous effluents, but a large amount of water is condensed following the shift converter and ahead of acid gas removal. This water will contain ammonia, H_2S and possibly small amounts of other materials such as cyanides, phenols, etc., if these are formed in gasification, or during startup or upsets. The sour water will have a very strong odor and care must be taken to avoid possible leaks or spills. Normally, it will all be returned to the process and used as part of the quench water ahead of the sand filters in order to dispose of it without causing emissions to the air. A nominal amount of sour water storage capacity would be desirable to assure that none will have to be discharged during start-up or upsets.

While not directly associated with effluents to the air, it should be pointed out that the closed system for handling sour water and disposing of it by total recycle may have to be modified. Compounds such as ammonia, phenols, etc., will be removed rather completely from the gas during condensation and recycled to the quench point. Since they are not destroyed in quenching, they will build-up in concentration in the circulating sour water stream, so that facilities may have to be added to separate them and purge them from the system. A similar situation can occur with volatile trace elements. This subject is discussed further in Section 6. EFFLUENTS - LIQUIDS AND SOLIDS.

5.5 Acid Gas Removal

This system removes sulfur compounds such as H_2S and COS as well as CO_2, using the proprietary Benfield process (14). It uses a hot solution of activated potassium carbonate in two separate absorber systems in series. The first of these produces an acid gas stream with a relatively high content of H_2S, which is sent to a Claus sulfur recovery plant. There should be no specific emissions to the air from this first scrubbing system.

In the second step residual sulfur is removed together with most of the CO_2, producing a gas stream which is discharged to the atmosphere. A further description of this acid gas removal operation is given in Reference (9), which points out that the sulfur content of this CO_2 stream is 3400 ppm and will require additional processing to clean it up before release to the atmosphere. One method is to use molecular sieves to adsorb the H_2S which is then desorbed and sent to the Claus plant, and this provision is included in our environmental study.

The use of molecular sieves was said to be quite expensive, but other techniques are available for consideration. One possibility is to use an absorption/oxidation type process to remove H_2S from the CO_2 vent stream. H_2S would be oxidized using an activated scrubbing liquid to form free sulfur which is separated as a by-product. Such processes are offered for commercial use by Stretford, IFP, and Takahax (15). Subsequent to the original study, Air Products indicated that the Rectisol process which uses methanol scrubbing at low temperature would be better

for acid gas removal in this application. However, results from plant operation (38) and from planning studies (37) show that the CO_2 vent stream from Rectisol also has an unacceptably high sulfur content. Moreover, it contains over 1 vol % combustibles including ethane, ethylene, methane, and carbon monoxide. Incineration of this vent gas is necessary from the standpoint of odors, combustibles, and H_2S content (3,5).

Since flue gas desulfurization is used elsewhere in the process, on the coal drier and utility boiler, it may be that this CO_2 stream could be blended in and cleaned up with incremental additions to the flue gas desulfurization system.

This particular BI-GAS design does not use air-fin cooling; instead, all of the waste heat is transferred to cooling water. However, in many applications the design will use air-fin cooling in order to minimize the load on the cooling tower and the water make-up requirement. In such cases careful consideration must be given to potential emissions to the air. With air-fin exchangers, a very large volume of air is passed over the exchanger surface, and in the event of leaks or tube failures, a considerable amount of material can be dispersed in the air, causing serious emissions to the atmosphere. This can be more of a problem for operations at very high pressure, as at 1000 psig, and on contaminated streams such as sour water.

The problem is not avoided by using cooling water, since any leakage will be into the cooling water which then flows through the cooling tower where it is efficiently stripped by a large volume of air.

5.6 Methanation and Drying

After acid gas removal, the gas is reheated, passed through a sulfur guard bed, and then to the methanation reactor. The system is all enclosed, hence there should be no major effluents to the air. However, there is considerable equipment that can be expected to contribute miscellaneous emissions, including:

- Exchangers that may leak or fail.
- Recycle gas compressors and valves.
- Circulating cooling water.

Leaks can be expected from such equipment operating at 1000 psig, especially from seals. Methods have been developed for estimating the amount of leakage in oil refineries, and techniques for monitoring and reducing emissions have been carefully considered (16). Such background should be applied in designing gasification plants so as to minimize potentially undesirable emissions.

Clean condensate is recovered after methanation, and when this is depressured, some gas will be released which should be recovered or incinerated. Water is also separated in the final glycol drying step, and should be recovered rather than being released to the atmosphere.

5.7 Auxiliary Facilities

One of the auxiliary facilities associated with the process is the Claus plant to recover sulfur. The acid gas containing sulfur compounds is first burned with added air to form free sulfur which is condensed and recovered. This is followed by additional stages using a catalyst to allow operating at lower temperature so as to increase the sulfur recovery. A typical value for sulfur recovery may be 97% in a three-stage operation, provided the feed gas contains 20% or more of H_2S. This would still give excessive sulfur emission in the Claus plant tail gas, amounting to about 25 tpd of SO_2 for this case. It is, therefore, necessary to add tail gas clean-up, and this modification has been included in our balances and calculation of thermal efficiency. A number of processes are offered commercially for such tail gas clean-up (17).

One other consideration on the sulfur plant is to control odor emissions due to leaks or associated with handling the product sulfur. There is an appreciable solubility of H_2S in molten sulfur, and it may escape during handling or storage; however, there are well established techniques for controling this and other possible sources of contamination such as sulfur dust.

The plant producing oxygen for gasification is relatively clean, and the major effluent to the air is waste nitrogen. The operation is conventional and is not expected to emit undesirable compounds or odors. It is, of course, a large energy consumer and so affects the size of the utilities system, and contributes significantly to the total amount of waste heat that must be dissipated from the process.

Perhaps the major source of contaminants emitted to the air is the utilities system which includes steam generation, power generation, cooling water, treating of make-up water and waste water, as well as miscellaneous items such as utility air and instrument air supplies. Coal is used as fuel in the boiler and steam superheater. It has a high sulfur content corresponding to 5.64 lb of SO_2 per MM Btu vs. an allowable value of 1.2 for large stationary boilers. Consequently, control measures such as flue gas clean-up will be needed. A sulfur removal of 80% would be sufficient, and this level of desulfurization has been achieved or exceeded by many of the processes offered for commercial use. In addition, control of fly ash emission is required when burning coal. For this case an ash removal of 98.16% is needed in order to meet the target of 0.1 lb of dust emission per MM Btu's. This level of removal has been obtained with flue gas scrubbing.

Instead of burning coal, it would be possible to use part of the low sulfur, low Btu gas made in the process as fuel in order to limit the emissions of sulfur and dust. This would consume a sizeable part of the total raw gas since the boiler fuel consumption corresponds to 16.4%

of the gas production, while the steam superheater consumes an additional 3.4%. There is also a loss in efficiency, since gasification to make low Btu gas has an estimated thermal efficiency of 77%, whereas flue gas desulfurization is indicated to have a considerably higher efficiency (95%). For any specific case, these alternatives need to be considered and evaluated, including credits for the gas fuel route which may result from using a combined cycle wherein the gas is first burned in a flue gas turbine to generate power, and is then used in a furnace for steam generation.

Emissions of NO_x must also be defined and controlled in a specific application of the process. The amount will depend on the furnace design, use of staged combustion, fuel nitrogen content, etc. In general, NO_x production can be decreased by designing for a lower flame temperature and by using low nitrogen fuel. Low Btu fuel gas is attractive from these standpoints. Processes are being developed to remove NO_x from flue gas, and a satisfactory progress will probably be available soon.

The flow rate and composition of the flue gases from the boiler for burning either coal or low Btu gas were compared in Table 4. Including the steam superheater furnace, the volume of flue gas from the utilities area is more than twice the volume of pipeline gas produced.

As is true for many other gasification processes, by far the largest effluent to the air is from the utility cooling tower. Flow of air through the cooling tower is 85,000 MM SCFD. In addition, there is a drift loss due to mist carried out by the air. A typical estimate of this would be about 263,000 lb/hr, although it could be reduced considerably by using some of the newer techniques that are being developed to control drift loss from cooling towers (18). Drift can cause deposits in the nearby area due to dissolved solids in the cooling water. Careful consideration should also be given to the potential fog problem or plume associated with cooling towers due to condensation under unfavorable atmospheric conditions. One way to avoid the plume is to provide reheat on the air leaving the cooling tower, but this will not normally be warranted. It may be that these problems can be taken care of by proper design and placement of the cooling tower.

Normally, there will not be contaminants introduced into the cooling water circuit that might be stripped out by the air flowing through the cooling tower. However, experience has shown that leaks can be expected in exchangers used in cooling water service, especially at high pressures such as the 1000 psig in this process. Leaks, for example, in exchangers on sour water service could introduce sulfur, cyanide and ammonia into the cooling water, which would then be stripped out into the air. Special precautions and possibly monitoring equipment may be needed from this standpoint.

The volume of air passing through the cooling tower is so large that every precaution should be taken to see that it does not inadvertently become contaminated. For this design, the air flow is about 85,000 MM CFD, or roughly 340 times the volume of pipeline gas produced, and is by far the largest gas stream released to the environment.

6. EFFLUENTS - LIQUIDS AND SOLIDS

Emissions to the environment of liquid and solid effluents will be discussed in the order in which they appear on the flow plan of Figure 1. Individual streams are identified on Figure 2 and described in Table 3.

6.1 Coal Preparation

A first and major effluent is the refuse from coal preparation and cleaning. This includes the rock and gangue delivered with the run of mine coal. Such refuse is separated and rejected after the first breaker, which crushes run of mine coal. Additional refuse is separated in the washing operation. These streams amount to 4,804 and 3,904 tpd respectively and will contain some coal as well as pyritic sulfur. They are therefore subject to oxidation and leaching, can cause pollution problems similar to acid mine water, and should be reviewed and considered from this standpoint (28).

The refuse might be returned to the mine or used as land fill provided the potential problems of secondary pollution are evaluated and controlled. The enormous magnitude of this effluent stream is illustrated by the fact that it amounts to over 860 acre feet per year of refuse to be disposed of. It is obvious that very careful and thorough planning will be necessary to avoid unexpected problems due to pollution from leaching of acid or soluble compounds and metals, or from dust.

In the washing operation, wash water will be sent to a settling pond where fines will be removed so that the water can be reused. Disposal of these fines, or tailings, must be provided for. Handling of the fines will call for special precautions, since if they are spilled on the ground they can dry out and then become dispersed by the wind or by trucks using the area. The system should be designed for complete recycle of the wash water so that there is no water effluent from the operation, which would present a difficult clean-up problem from the standpoint of dissolved and suspended materials.

Leaching, or seepage, through the bottom of the tailing pond should also be controlled. In a heavy clay-type soil this may not be a problem; however, in sandy soil it may be necessary to provide a barrier which might be a layer of plastic or clay.

A further consideration on the coal preparation area is with regard to the coal storage pile. The design includes 30 days' storage, or about 700,000 tons; so the coal storage pile will cover a very large area. Rain runoff can lead to undesirable effluents. A large part of the rain can run off quickly and carry suspended particles, while the remainder will have a long contact time with the coal and can pick up acids and organics. Therefore, rain runoff from the storage area should be collected in storm sewers and sent to a separate storm pond. With a certain amount of treatment, this water can then be used as make-up for the process. Control of seepage may be desirable on the pond, and particularly on the coal storage area, using for example, a layer of concrete, plastic or clay.

Other effluents from the coal preparation area are associated with coal drying. Coal fines that are picked up by the drying gas should be recovered by bag filters or scrubbers and returned to the process. In addition, where coal is used as fuel on the dryer there will be by-product ash to be recovered and disposed of. It might be included along with the refuse from coal cleaning, or it could be combined with the slag from gasification and disposed of as land fill.

6.2 Gasification

Coal feed is reacted with steam and oxygen at high temperature and about 80 atmospheres pressure in the gasifier. The major effluent from this section is the slag formed from ash in the coal. Essentially, all of the ash in the feed is rejected here, after having been fused in the lower zone of the gasifier which operates at 3000°F. Molten slag is quenched in water and thereby shattered to form a slurry which is then depressured using lock hoppers for removal from the high pressure system. The slag should be relatively sulfur free and unreactive, having been fused at high temperature. Also, it contains little or no carbon and therefore can be discarded. For handling, it can be mixed with an equal weight of water to form a slurry. This water will pick up dust from the slag, and can leach out soluble salts and metals; therefore, it should be collected and reused so as not to become an effluent from the plant. The ash slurry might be dewatered for disposal in the mine and the water sent to a holding pond for reuse.

Production of dry slag is 68,391 lb/hr, corresponding to about 90 acre-ft. per year; consequently, adequate provision for disposal is needed. The other major stream leaving the gasifier is the raw gas product. It contains a large amount of char which is blown out of the gasifier and recovered in cyclones for recycling to the lower zone of the gasifier. No other streams are normally released to the environment from the gasification section.

6.3 Quench and Dust Removal

A large part of the coal feed to the gasifier is blown overhead since the reactor operates at high velocity and with high entrainment. This char is separated in a cyclone where quench water is introduced. The recovered char goes to lock hoppers and a feeder which returns it to the lower zone of the gasifier. Except for leaks and maintenance, there should be no emissions from these facilities.

The raw gas is further quenched with sour water in a quench vessel ahead of the sand bed filters. These filters operate in parallel, and when the pressure drop builds up, one unit is cleaned by back blowing. Dusty gas from the backblowing operation is returned to the gasifier. The cleaned sand filter is then placed back in service.

An important feature of the sand bed filters is that they provide dust removal at high temperature, such that water does not have to be condensed. This is a major advantage when using a sulfur resistant shift catalyst so that shift conversion is carried out before the raw gas has been cooled and the moisture condensed out. Thus, the steam required for shift conversion is provided partly by residual steam leaving the gasifier, together with sour water which is introduced as quench. This arrangement provides a convenient and effective way to dispose of sour water.

Ash removed by the sand filters will be returned to the lower gasification zone and can leave with the slag. However, there will be some volatile components, such as arsenic and zinc compounds etc., that will be revaporized and carried up with the gas. They can then condense again and be caught by the sand bed filters. It will be seen that this constitutes a system with total recycle, with no way for certain materials to escape. Therefore, it may be necessary to provide a purge stream in order to remove such materials. For example, part of the dust recovered by the sand bed filters could be removed for disposal. The composition and nature of this stream cannot be estimated at this time, neither can the amount be predicted. The required information should be obtained during operation of the pilot facilities.

Similarly, it may be necessary to provide a purge stream, or separation system, on the sour water if certain chemical compounds or trace elements tend to recycle and build up in concentration.

6.4 Shift Conversion

As pointed out, shift conversion is carried out before sulfur has been removed from the raw gas. Therefore, a sulfur resistant shift catalyst is required, and these normally have lower activity than catalysts which are used on sulfur free gas. Steam in the entering gas is adjusted to give about one mole per mole of dry gas. Steam conversion in the shift reactor is about 27%, while 62% of the CO entering is reacted.

After shifting, the gas is cooled to condense out most of the remaining moisture, which gives 866,613 lb/hr of sour condensate. This water will contain H_2S and other sulfur compounds as well as ammonia and probably traces of phenols, cyanides, etc. that are present in the gas. This sour water can all be disposed of by recycling to the process to provide part of the quench required at the outlet of the gasifier.

In this particular BI-GAS design, the amount of water consumed in the shift reaction is 385,630 pounds per hour so that this much unreacted water in the gases leaving the gasifier could be disposed of without having a net production of sour water from the process. In addition there is 68,270 pounds per hour of water used for quenching the slag, and perhaps this could be an additional consumption of sour water.

It should be pointed out that compounds such as ammonia and phenol will dissolve in the sour water and be recycled through the quench. Of course, quenching does not actually destroy these materials, although they might be destroyed in the shift reactor, and this is a distinct possibility which would be worthwhile to explore. If they cannot be destroyed then it will be necessary to provide a purge stream, which is further processed to separate compounds that build up in the recycle stream. For example, ammonia and phenol could be separated and taken off as by-products for sale or for incineration. Trace elements that are volatilized in the gasifier, such as arsenic, boron, lead, etc., may also tend to build-up in the circulating sour water stream and have to be removed and disposed of. This subject is discussed further in Section 10 - TRACE ELEMENTS.

One possible modification is to provide stripping on a portion of the recycle sour water stream so as to remove volatile materials such as ammonia. In the case of less volatile soluble materials such as phenols, these will tend to build up and it may be necessary to add an extraction step to separate them and remove them from the system. It is possible that contaminants may be destroyed in the shift conversion reactor by hydrogenation as a result of the large amount of hydrogen present. Further exploration of this possibility would be desirable. Oxygenated compounds might also be destroyed, and perhaps the rate of the ammonia equilibration reaction would be sufficient to control the concentration of ammonia to an acceptable level. On the other hand, such recycling might undesirably increase the concentration of some materials such as cyanides and thiocyanates. Obviously, more information is required to define the situation.

While this particular design of the BI-GAS process does not show liquid or solid effluents or by-products from this section of the plant, further clarification and information is needed from pilot plant operations regarding contaminants such as ammonia, cyanides, phenols, etc., that may be formed in gasification and tend to concentrate in the recirculated sour water. They will either have to be destroyed in the recycling operation or removed from the system by using appropriate separation techniques. Ammonia is of particular concern since in many gasification processes about 60% of the nitrogen in the coal is converted to ammonia. It is relatively easy to separate and remove as a valuable by-product, and for this design the production of ammonia could be of the order of 100 tons/day.

Other trace materials may be much more difficult to separate and dispose of. For example, it is known that many trace elements will volatilize to a considerable extent during gasification. Such elements include mercury, arsenic, antimony, cadmium, zinc, fluorine, boron etc. and many of these can be quite toxic. To some extent, they may be removed by the sand filters and thereby returned to the gasifier. However, it is unlikely that they will leave with the molten slag, and therefore may recycle between the gasifier and sand filters and build up in concentration.

If this happens, perhaps they could be removed by purging some of the dust recovered on the sand filters to a separate metal recovery system. It is also likely that some of these metals will pass through the sand filters and show up in the sour water stream. Since this is also recycled completely, there is no place for such metals to leave the system; consequently, they will build-up in concentration in the recycled sour water stream. Again, it may be that part of this stream could be processed to separate and recover such materials.

In any event, it is apparent that provisions will have to be made for removing from the system materials such as trace elements that are volatilized in the gasifier. It is most important to obtain the additional information needed in this area to define the problem and proper controls. The amount and nature of the trace elements leaving the gasifier should be carefully determined during pilot plant operation, so that environmental aspects can then be properly evaluated. This is one area where additional information is urgently needed.

6.5 Acid Gas Removal

The acid gas removal system is intended to remove sulfur compounds as well as CO_2, prior to methanation. Amine scrubbing is commonly used for this purpose but is not effective on removing forms of sulfur other than H_2S, such as COS and CS_2 which may be present. Other techniques may use hot potassium carbonate scrubbing or absorption with refrigerated methanol both of which are effective for removing carbonyl sulfide. Another route is to use absorption/oxidation systems where the H_2S is reacted directly to free sulfur, which is then separated as a by-product. This type of system is offered by Stretford, Takahax, IFP and others, but may not give adequate removal of COS, etc. Of course, a separate system is then needed for CO_2 removal.

This particular BI-GAS design uses the Benfield hot carbonate system to provide two separate gas streams. One of these is relatively high in H_2S content for processing in the sulfur plant, while the other is a CO_2 stream relatively low in sulfur to be vented. There are no major liquid or solid effluents from this operation; however, it is necessary to purge a small amount of the scrubbing solution since certain contaminants build-up and interfere with the operation. The amount and composition of this purge have not been given, but it probably contains an appreciable amount of potassium carbonate, and might be disposed of by neutralizing it with sulfuric acid that is used in the water treating system for regenerating ion exchange residence. It might also be processed for recovery. Some suitable disposal needs to be defined.

One complication that occurs in all processes where coal is gasified with oxygen results from the formation of carbonyl sulfide in the gasifier. It generally results in complications and limits the choice of processes for acid gas removal, since carbonyl sulfide is not removed adequately by conventional amine scrubbing. An interesting possibility is to react COS with steam over a catalyst at moderate temperatures so as to convert it to H_2S, which can readily be removed. It has been shown that catalysts such as alumina will promote this reaction (19). Possibly, COS conversion could also be carried out in the shift reactor or in a separate bed of suitable catalyst.

The scrubbing towers of the acid gas removal system will no doubt be very effective for removing trace amounts of dust or other materials which have not been separated by the upstream processing. Such materials will also accumulate in the scrubbing solution and may have to be separated and purged. In some other processing schemes, filtering of the scrubbing solution has been included to separate solid particles which are then rejected from the system. Depending on the nature of such materials they might be disposed of along with the slag from gasification or possibly processed for recovery.

Again, it should be pointed out that certain trace elements will be volatile to some extent in the gasifier and will be carried out in the raw gas. These must show up in the downstream processing, where they will be separated out. The amounts can be very significant. For example, a concentration of only 10 parts per million in the entering coal corresponds to a total of 240 lb/day, a large part of which may volatilize in the gasifier. Since there are a large number of elements to consider, the total amounts to be disposed of can be very formidable, particularly if they are toxic, as is the case for many volatile elements. Information is needed on where they will appear, and in what form, so that the situation can be evaluated and proper control measures included as required.

.6.6 Methanation and Drying

Following acid gas removal, the gas is quite clean and should not contain significant amounts of undesirable contaminants. Methanation produces 214,818 lb/hr of water, which is condensed and is suitable for boiler feed water make-up. The large heat release in the methanation reaction is used to generate high pressure steam, but this is used within the process and is not an effluent from the plant. Finally, the gas is dried with glycol to meet pipeline specifications. Water removed at this point is 27,219 lb/hr and, again, should be suitable for make-up water if it is recovered. There may also be a small amount of purge from the system containing glycol, which could be incinerated or passed through the biox system for clean-up.

6.7 Auxiliary Facilities

As previously discussed, there will be solid effluents from the furnaces burning coal for utilities production. Residual ash can be disposed of together with the slag from gasification. In addition, there is spent limestone from flue gas desulfurization which can be similarly handled and also sludge from water treating. Sludge from biological oxidation should be incinerated to avoid odor problems.

Some water must be purged from the cooling water system in order to control concentration of dissolved solids. This represents the minimum net discharge of water from the plant. While there will also be blowdown from boilers, it can be used as cooling tower makeup, and the sour water will be cleaned up for reuse. Water discharged from the plant will contain sodium chloride, sulfates, and other dissolved solids. The amount, composition, and disposition need to be carefully defined and evaluated in any large scale application.

7. GAS HANDLING AND SOUR WATER CONSIDERATIONS.

There are a number of possible ways which can be used to clean-up the raw gas leaving the gasifier so that it can be methanated to high Btu pipeline gas. The choice can have a very large effect on effluents and particularly on the production of sour water and the disposal of it. To a large extent, the choice is a matter of selecting the order in which the processing steps are carried out, and some of the routes are as follows:

(1) The hot gas goes first to shift conversion using a sulfur resistant catalyst, followed by acid gas removal and methanation.

(2) The raw gas is cooled and scrubbed for acid gas removal, and then goes to shift, CO_2 removal, and methanation.

(3) The gas is desulfurized at high temperature such as 700°F instead of using amine or hot carbonate scrubbing, and is then methanated directly with steam, followed by CO_2 removal.

The first of these is the route usually planned in making SNG by coal gasification. It requires gas clean-up ahead of the shift reactor in order to remove materials that might foul the shift catalyst, particularly dust and tar. Catalyst activity is relatively low due to the presence of sulfur and a large excess of steam is generally needed to control deposits on the catalysts.

The second type of system is more typical of operation to manufacture high purity hydrogen, where the shift reaction must be maximized. An active shift catalyst can be used since it is not exposed to sulfur. However, if this route is used in making SNG, the gas must be cooled after shifting in order to scrub out CO_2 before methanation. This introduces an extra heating and cooling step which is inefficient. Thus, the entire gas stream is cooled three times compared to twice in the first case.

The third approach is a new proposal for gas processing which should be simpler and more efficient, although it requires development of technology. With this combination the gas is only cooled once, and that is after the final methanation. The gas is first desulfurized at high temperature using a process such as that studied by the Bureau of Mines based on the reaction of iron with sulfur (30), or that studied by CONSOL based on half-calcined dolomite. Next, the CO is methanated directly by reaction with steam rather than with hydrogen. It should be possible to react CO with steam to form methane and CO_2, since this type of reaction is carried out in a number of processes making SNG from naphtha by reacting it with steam to form methane and CO_2 (31,32,33).

It would also be possible to use conventional shifting and methanation in two separate stages, but it is more efficient to react the CO to methane directly since it requires less steam. In other words, it combines the steam consumption of the shift reaction together with the steam formation in methanation, whereby the actual steam requirement is reduced.

With this route it will be necessary to remove sulfur, dust and tar at high temperature ahead of methanation. As mentioned, sulfur might be removed in a bed of iron type adsorbent, which might also remove dust, or the dust could be removed in sand bed filters. Tar could be removed by scrubbing, for example, at 500-600°F. While the projected methanation reaction has been used effectively on naphthas, it will probably be more difficult to obtain the desired conversion on heavier oils or on aromatic type compounds. Therefore, the system may be best suited for those gasification processes that do not make tar or naphtha.

An advantage for removing CO_2 after methanation is that there is less volume of gas and the concentration of CO_2 is higher. Of course, the total amount of CO_2 to be removed is the same as in the other routes.

It should be pointed out that compounds such as phenol and ammonia in the raw gas can pass through the system and may not be removed until the final cooling step. In addition, the effect of volatile trace metals must be considered. It is not clear whether these would be removed along with the dust or whether they might deposit on the catalyst and affect its activity.

The potential savings and simplification possible with this modified system for gas cleaning would seem to provide considerable incentive to develop suitable techniques for removing sulfur and dust at high temperatures. Techniques are known for removing small amounts of sulfur at high temperature, for example, using iron, zinc oxide, or nickel base materials. The problem has been that these cannot be conveniently regenerated, and therefore are not practical for removing large amounts of sulfur. It should be possible to develop practical regeneration techniques, so that the sulfur adsorbent could be recirculated and used continuously or batch wise.

Dolomite may also be a promising prospect for such a system based on background available from the CO_2 Acceptor process development (4). This work has shown that dolomite will remove sulfur compound from gases at high temperature. It has also been shown that the spent material can be desulfurized and regenerated by reacting with CO_2 in a water slurry at 190 F to produce a stream of H_2S which is available at a reasonably high concentration so that it can be processed efficiently in a conventional Claus plant.

If techniques were developed for removing dust and sulfur at high temperature, then they would also be useful for making clean, low Btu fuel gas from coal. For example, coal could be gasified with air or oxygen, and after clean-up used in process furnaces or utility boilers, which would then not require individual stack gas clean-up. The system should have a higher thermal efficiency than conventional systems to make

low Btu gas in that little or no sensible heat in the hot gas from gasification would be wasted. A further very important advantage is that the clean fuel gas could be used in a combined cycle for power generation. That is, the low Btu gas would first be used in a gas turbine generating electric power, and it would then go to a furnace for final combustion and steam generation.

8. SULFUR BALANCE

Details on the amount of sulfur in the various streams entering and leaving the plant are shown in Table 5. Essentially all of the sulfur emission from the plant is to the air, and most of this is in the flue gas discharged from the steam boiler and super heater. Of the sulfur entering the plant in the coal feed, 83.4% is recovered as by-product sulfur from the sulfur plant. An additional 12.4% of the sulfur is removed and rejected by the flue gas desulfurization facilities on the utilities furnaces and coal dryer. A number of processes are offered for stack gas cleanup, and many of these can give a sulfur removal well above the 79% target, at little or no added cost.

Streams such as the waste water discharge and the CO_2 vent gas will be cleaned up to avoid odor problems, and will then contain negligible amounts of sulfur. Thus, the raw CO_2 stream from acid gas removal contains 3400 ppm H_2S which is removed by molecular sieve adsorption and sent to the sulfur plant. Similarly, the slag is assumed to be free of sulfur and not a contributor to pollution. These items should be examined carefully in a final plant design. While the gas liquor contains considerable H_2S, most of this will be removed in the sour water stripper and sent to the sulfur plant. The biox unit provides final cleanup on the effluent water.

Table 5

Sulfur Balance (1)

Sulfur Input in Coal (1)	lb/hr	%
To gasifier	35,487	84.2
To coal dryer	418	1.0
To utility steam boiler	5,164	12.2
To steam superheater	1,086	2.6
TOTAL IN	42,155	100.0
Sulfur Output (1)		
From Claus plant (2)	35,132	83.4
In Claus tail gas (2)	355	0.8
In pipeline gas product	nil	---
In treated sour water	nil	---
In CO_2 vent gas	nil	---
In slag	nil	---
From flue gas desulfurization coal dryer (3)	330	0.8
From flue gas desulfurization boiler (3)	4,080	9.6
From flue gas desulfurization superheater (3)	858	2.0
In flue gas on coal dryer (3)	88	0.2
In flue gas on boiler (3)	1,084	2.6
In flue gas on superheater (3)	228	0.6
TOTAL OUT	42,155	100.0

NOTES:

(1) Does not include refuse from coal cleaning operations which could be sizable and needs to be defined.

(2) Basis: 99% S recovery including tail gas clean-up.

(3) Based on flue gas desulfurization with 79% S removal.

9. THERMAL EFFICIENCY

The base thermal efficiency for the process is obtained by comparing the heating value of the net pipeline gas produced, with that for the total coal used including gasification and all utilities production. As shown in Table 6, the base efficiency is 65.9%. Coal is used as fuel in the coal drier and the utilities systems, requiring flue gas clean-up; but, the fuel required for this is not included in the above number. However, it is estimated that this will increase the fuel requirements by less than 5% on the individual furnaces, and less than 1% on the total coal delivered to the plant. Base efficiency with this allowance will then be about 65.3%

An alternative would be to use low sulfur low Btu gas made in the process as fuel to the furnaces. Pollution due to sulfur and ash would then be avoided, but it would still be necessary to provide good dust removal on the coal dryer. Obviously, such use of gas fuel will appreciably reduce the amount of gas available to the pipeline, and correspondingly decrease thermal efficiency of the process, as illustrated below:

Fuel used in:				
Coal dryer	coal	gas	gas	gas
Steam superheater	coal	coal	gas	gas
Utility boiler	coal	coal	coal	gas
MM Btu/hr in:				
Coal consumed	14,920	14,775	14,390	12,560
Low Btu gas	11,191	11,046	10,661	8,831
Pipeline gas	9,830	9,703	9,365	7,757
Thermal Efficiency %	65.3	65.1	64.6	61.8

Using all gas fuel instead of coal decreases thermal efficiency from 65.3% to 61.8%. At the same time, production of pipeline gas is reduced by 21% for a given size of gasifier.

These results again emphasize the desirability of applying an efficient flue gas clean-up operation, so as to allow using high sulfur coal as fuel.

It is also of interest to look at the thermal efficiency for this specific design as a way to make low sulfur, low Btu gas. Heating value in the gases prior to methanation corresponds to about 74% thermal efficiency for the gasification step, including an allowance for flue gas clean-up. Of the total heating value in the low Btu gas, 41.6% is contributed by its methane content.

In considering thermal efficiency of the process as a source of clean low Btu fuel gas, it is proper to exclude the shift conversion operation since it is not needed. Then the quenching can be omitted and replaced with a more efficient heat exchanger to generate high pressure steam, thereby decreasing the size of the utility steam boiler. This reduces the total coal consumption and adds another 3% to the thermal efficiency, bringing it up to 77% for making clean fuel gas.

Thermal efficiencies for the various alternatives considered in the BI-GAS process are summarized in Table 6.

Table 6

Thermal Efficiencies for BI-GAS Process

Base Coal * Consumption	lb/hr	MM Btu/hr
Gasifier	1,018,700	12,560
Coal dryer	12,000	145
Steam Superheater	31,200	385
Utility boiler	148,400	1,830
	1,210,300	14,920

Base Case Thermal efficiency	% Efficiency
Without flue gas desulfurization	65.9
With flue gas desulfurization	65.3
With low Btu gas fuel to:	
Coal dryer	65.1
and steam superheater	64.6
and utility boiler	61.8
Alternative to make only low Btu gas	
Base case design	74.0
Without shift conversion	77.0

* Based on 8.4% moisture

10. TRACE ELEMENTS

Coal contains many trace elements present in less than 1% concentration that need to be carefully considered from the standpoint of potential impact on the environment. Many of these may volatilize to a small or large extent during processing, and many of the volatile components can be highly toxic. This is especially true for mercury, selenium, arsenic, molybdenum, lead, cadmium, beryllium and fluorine. The fate of trace elements in coal conversion operations, such as gasification or liquefaction, can be very different than experienced in conventional coal fired furnaces. One reason is that the conversion operations take place in a reducing atmosphere, whereas in combustion the conditions are always oxidizing. This maintains the trace elements in an oxidized condition such that they may have more tendency to combine or dissolve in the major ash components such as silica and alumina. On the other hand, the reducing atmosphere present in coal conversion may form compounds such as hydrides, carbonyls or sulfides which may be more volatile. Studies on coal fired furnaces have indicated that smaller particles in fly ash contain a higher concentration of trace elements, presumably due to volatilization of these elements in the combustion zone and their subsequent condensation and collection on the fly ash particles (20). Other studies on coal fired furnaces are pertinent (21,22,23) and some of these report mass balances on trace elements around the furnaces (24).

Considerable information is available on the analyses of coal, including trace constituents, and these data have been assembled and evaluated (25,26). A few studies have been made to determine what happens to various trace elements during gasification (2,27). As expected these show a very appreciable amount of volatilization on certain elements. As an order of magnitude, using the factors for this specific BI-GAS design would result in 240 lb/day carried out by the gas for each 10 ppm of trace element volatilized from the coal.

In order to make the picture on trace metals more meaningful, the approximate degree of volatilization shown for various elements has been combined with their corresponding concentration in a hypothetical coal (as typical), giving an estimate of the pounds per day of each element that might be carried out with the hot gases leaving the gasifier. Results are shown in Table 7 in the order of decreasing volatility. Looking at the estimated amounts that may be carried overhead, it becomes immediately apparent that there can be a very real problem. For each element the net amount carried overhead must be collected, removed from the system, and dispose of in an acceptable manner. In the case of zinc, boron and fluorine the degree of volatilization has not yet been determined, but they would be expected to be rather volatile. Even if only 10% of the total amount is volatile, there will be large quantities to remove in the gas cleaning operation and to dispose of.

Table 7

Example of Trace Elements That May Appear in Gas Cleaning Section

Element	Possible ppm in Coal (a)	% Volatile for example (b)	In Gas lb/day (c)
Cl	1500	>90+	32,400
Hg	0.2	90+	5
Se	2.2	74	39
As	31	65	484
Pb	7.7	63	116
Cd	0.14	62	2
Sb	0.15	33	1
V	35	30	252
Ni	14	24	81
Be	2	18	9
Zn	44	(10)	106
B	165	(10)	396
F	85	(10)	204
Ti	340	(10)	816
Cr	22	nil	nil

(a) Mainly based on Pittsburgh Seam Coal (2).

(b) Mainly based on lower temp gasifier (27) and indicated at 10% for Zn, B, and F, in absence of data.

(c) For 12,000 tons/day of coal feed

A complication results in the gas clean-up section due to the presence of volatile trace elements. In the BI-GAS design, raw gas from the gasifier is cooled and cleaned to remove all dust and other contaminants except for the more volatile ones such as H_2S. Contaminants collect in the dust and sour water both of which are returned to the system and presumably recycled to extinction. Chemical compounds such as cyanide may thereby be destroyed but this cannot be the case for trace elements. It will be difficult for volatile elements to leave with the slag through the 3000°F zone, they will therefore build up in the recirculating streams and have to be purged from the system.

The preceding discussion has been directed primarily at trace elements that are partially volatilized during gasification and that therefore must be recovered and disposed of in the gas cleaning section. Consideration must also be given to trace metals that are not volatilized and leave in the solid effluents from the plant, one of which is the slag from gasification. Undesirable elements might be leached out of this slag since it is handled as a water slurry and will ultimately be exposed to leaching by ground water when it is disposed of as land fill or to the mine. Sufficient information is not now available to evaluate the potential problems and the situation may be quite different from the slag rejected from coal fired furnaces since the slag is produced in a reducing atmosphere rather than an oxidizing one. Background information on slag from blast furnaces used in the steel industry may be pertinent from this standpoint, since the blast furnace operates with a reducing atmosphere. However, a large amount of limestone is also added to the blast furnace, consequently the nature of the slag will be different.

An additional source of possible contamination from trace elements is associated with the disposal of refuse from coal cleaning. It is known that contained sulfur compounds will oxidize upon exposure to the air and form an acid solution in the presence of water. It is quite likely that a number of trace elements can be extracted from the refuse by this acid solution. For example, similar systems have been proposed and studied for recovering copper, nickel, iron, etc., from low grade ores. It might be thought that this situation is no worse than that existing for natural mineral deposits; however, the conditions are quite different. First, the mineral has been crushed and reduced in size so that vastly more surface is exposed and available for extraction. In addition, the mineral is exposed to a large amount of oxygen, which together with the large surface area can cause considerable oxidation of sulfur compounds, organic materials, and minerals in the refuse, whereas natural mineral deposits are not subject to such conditions. Some studies have been made in this general area (28,29), but much more work is needed.

11. ALTERNATIVES TO CONSIDER

This section of the report covers various modifications to the base design that warrant further consideration and evaluation. Some of these are added as required to control pollution. Other alternatives are discussed that may improve thermal efficiency of the process, some of which will require additional experimental work or development of new technology. Table 8 summarizes a number of items. The first modification to consider is the addition of facilities to control sulfur emission on furnaces firing high sulfur coal as fuel.

In the base design, high sulfur coal is used for fuel on the coal dryer and in the utilities furnaces. Flue gas clean-up is therefore needed on each of these, and a large number of processes are offered commercially for this purpose (10,13,34,35). Some of these use scrubbing with lime, limestone, magnesia, or sodium carbonate solutions. In the case of limestone, the spent material is discarded, whereas in most other processes the scrubbing medium is regenerated to produce a by product sulfur compound such as sulfur, sulfuric acid, or gypsum. Those processes that use scrubbing will also remove particulates, as is required when burning coal as fuel.

Instead of using coal with flue gas clean-up, it would be possible to use part of the clean, low Btu gas produced in the process as fuel for the furnaces. In evaluating this route, allowance should be made for the large difference in thermal efficiency compared to burning coal directly. Some energy is required to operate flue gas desulfurization, such that the thermal efficiency corresponds to about 95%, whereas gasification to make low Btu clean fuel gas has a thermal efficiency of about 75%. This specific BI-GAS plant design used about 15.8% of the total coal feed for firing in the coal dryer and in the utilities area. Therefore, the energy for flue gas desulfurization will decrease overall plant thermal efficiency by about 0.6%, while the gasification route reduces it by about 3.0%.

In any event, the coal dryer will require dust recovery. Fortunately, fuel consumption for coal drying is small relative to the utility furnaces, so it is reasonable to use low Btu, clean gas as fuel on the coal dryer. One advantage is that this avoids complications that may result if coal is used as fuel, and the ash from this coal mixes with and contaminates the coal fines picked up by the gas in the dryer.

A second modification to consider is on the system for recovering and handling dust carried out by the raw gas leaving the gasifier. In the base design a large amount of char is entrained from the reactor and separated in a cyclone for return to the lower zone. Lock hoppers are used for repressuring the recovered char, and since these require the use of mechanical valves, the 1700°F raw gas and char are cooled to 1115°F by injecting quench water into the cyclone. Recovered char is then injected into the lower gasification zone which operates at 3000°F. It would of course be more efficient to return the char without cooling, but since mechanical valves are used, some cooling is necessary. However, there is an alternative to

Table 8

Alternatives to Consider

- Add stack gas clean-up to remove sulfur and dust on coal fired furnaces, or use low Btu, clean gas fuel.

- Omit cooling on cyclone after gasifier and use standpipe to return hot char to gasifier at 1700°F instead of 1115°F.

- Design coal dryer for coal fuel and low excess air (e.g. 10%) with vent gas recycle, to minimize fuel consumption and volume of vent gas to dust removal.

- Pressurize coal feed by pumping a water slurry, which is evaporated in a fluid bed using indirect heating, to form steam which is used in gasifier and shift convertor, while at the same time preheating the coal feed to 500-550°F.

  Evaluate alternate of using light hydrocarbon instead of water to make slurry. Hydrocarbon would then be condensed and reused.

- Remove excessive sulfur from CO_2 vent stream by using absorption/ oxidation process instead of molecular sieves.

- Instead of cooling to scrub out acid gases, remove sulfur at high temperature and then react CO with steam catalytically to form methane directly. Dolomite or iron system might be effective for sulfur removal. Final step is then cooling and CO_2 removal from smaller volume of gas.

- General efficiency items to conserve fuel:

  - heat pumps on acid gas removal and sour water stripper
  - air-fin exchangers to save cooling water
  - air preheat on furnaces

using lock hoppers which would not require cooling, in which the char would be recovered in cyclones at 1700°F and flow down through vertical standpipes to build up pressure on the fluidized char, as needed in order to return it to the lower zone. This general technique is well established for use in many fluidized solids operations, such as fluid catalytic cracking in oil refineries. Returning the hot char without cooling significantly reduces the heat load on the gasification zone, so that oxygen requirement is decreased by about 15%. This also reduces the gas volume to be handled, and the amount of CO_2 to be removed in the acid gas removal section.

Another modification that could improve efficiency of the process and simplify the operation is in the method of pressurizing the coal feed. It was originally proposed to use a piston feeder on the dry coal powder, since this inherently has minimum power consumption. At the same time it was suggested that lock hoppers could be used with pressure staging to minimize the amount of vent gas that would have to be collected and compressed. Lock hoppers are used commercially but are expensive, and the cyclic operation of valves requires considerable maintenance.

It has also been proposed to mix the coal with a liquid to form a slurry which then can be pumped into the high pressure system where the liquid is evaporated. The liquid may be water in an amount approximately equal to the weight of coal so that the slurry can be handled and pumped. In some cases a light hydrocarbon such as naphtha is used instead of water so as to reduce the heat load, in which case the vapors can be condensed and reused. Latent heat to evaporate naphtha is about 150 Btu per pound, compared to roughly 700 for water at high pressure.

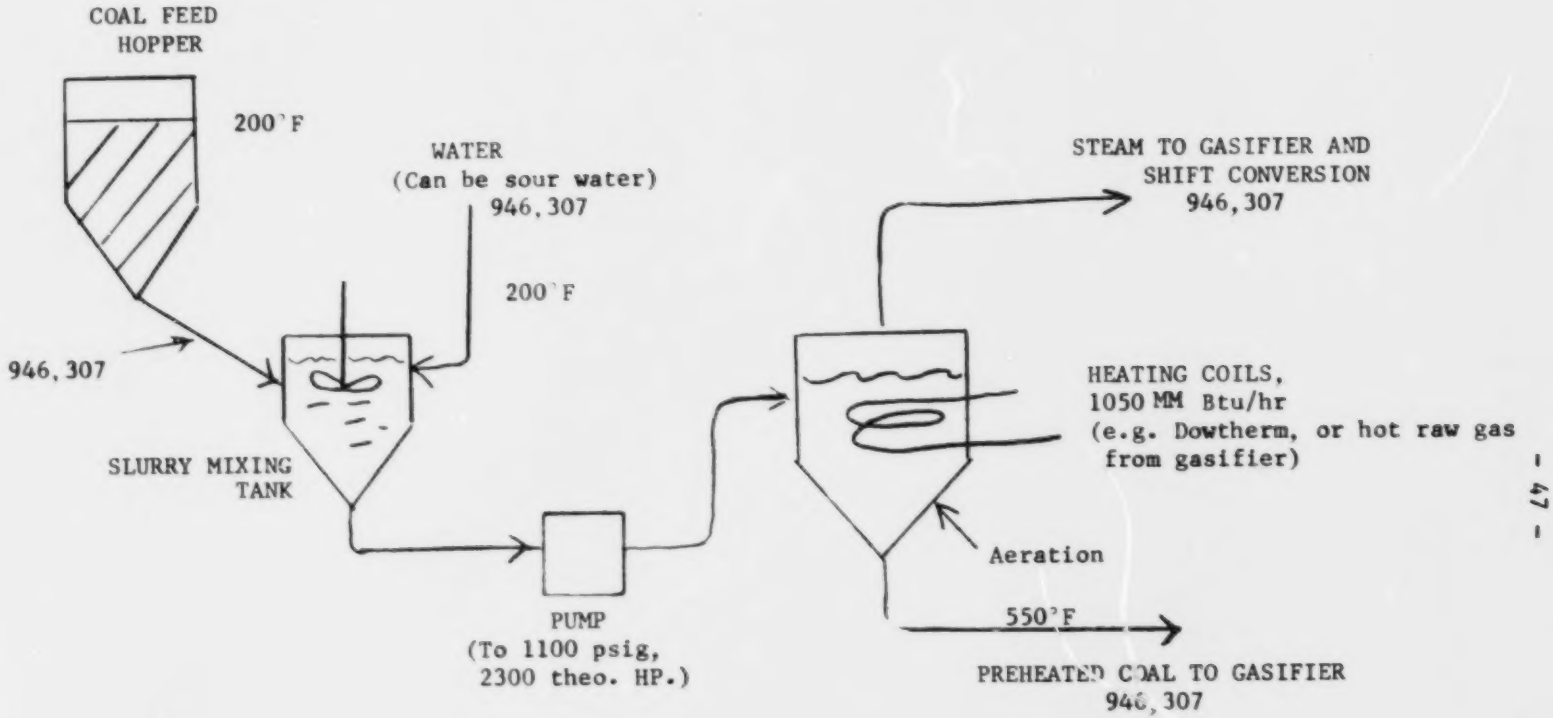
This route is promising, especially with water, since large quantities of steam are used in the gasifier and in shift conversion. The combined steam consumption is about 1.7 million pounds per hour, and heat for generating this steam must come either from waste heat or from furnaces. The quantity of steam is large relative to the coal feed rate of slightly less than 1,000,000 pounds per hour, and would be sufficient to form a pumpable slurry for feeding. If the coal feed were slurried with an equal weight of water and pumped to about 1100 psig, then it could be evaporated by indirect heat exchange with the hot raw gas available at 1700°F from the gasifier, after the char has been separated by means of a cyclone as previously described. Latent heat to evaporate the water amounts to about 600 million Btu per hour, which could be provided by sensible heat of the raw gas if it were cooled from 1700 to 900°F. It is therefore of interest to consider flowing the hot gas through a heat exchanger which would transfer heat to the coal slurry in order to evaporate it to dryness. The operating temperature would be about 550°F. and past work has shown that coal can be preheated to this temperature without evolving a large amount of volatiles or becoming plastic and sticky.

It may not be practical to carry out such an operation in a conventional tubular exchanger because of local overheating or plugging, but one possibility is to use a fluid bed in which there are heat transfer tubes, as proposed for fluid bed boilers. Such an approach is illustrated in Figure 3.

A water slurry is injected into the fluidized bed where it is evaporated to dryness. Similar systems have been used successfully on small commercial

Figure 3

POSSIBLE SYSTEM FOR PUMPING COAL TO HIGH PRESSURE, USING SLURRY



Numbers are flow rate, lb/hr.

units to evaporate various slurries. Heat is supplied indirectly by exchanger coils submerged in the bed, which are found to provide excellent heat transfer without fouling. Hot gases from the gasifier could flow through the coils to supply necessary heat. Instead of this arrangement, heat could be supplied from a furnace using for example Dowtherm or liquid metal to assure good heat transfer and temperature control.

Steam from evaporation of the slurry can be fed to the upper and lower zones of the gasifier as desired. Part of the steam can also be used in the shift converter. Although the steam will contain some solid particles, this should not matter. Dry coal is withdrawn from the bed and fed to the gasifier, for example, using a standpipe or screw feeder arrangement. It will be noted that the coal is preheated to 550°F in this system. Its sensible heat content is thereby increased by about 150 million Btus per hour, giving a corresponding saving in the overall heat load on the gasifier.

The base design uses hot carbonate scrubbing for acid gas removal and it has the advantage that it will remove carbonyl sulfide whereas amine scrubbing is not effective. Of the total sulfur in the raw gas some 10% of it may be present in forms other than H_2S, such as carbonyl sulfide, consequently these other forms of sulfur must also be removed in order to give satisfactory cleanup. This particular hot carbonate system makes a vent stream of CO_2 which is excessively high in sulfur content and needs to be cleaned up. Therefore we have added a molecular sieve unit to remove H_2S from this CO_2 stream and sent it to the sulfur plant for recovery. Further consideration should be given to catalytic hydrolysis of carbonyl sulfide to form H_2S. This would be used prior to acid gas removal, and might be combined with the shift operation as suggested by Bituminous Coal Research (36).

Acid gas removal is a large consumer of utilities, equivalent to about 10% on overall thermal efficiency. It therefore warrants thorough consideration of alternatives in order to arrive at an optimum system. In this connection, the desirability of hydrolysing carbonyl sulfide prior to acid gas removal should be emphasized. If this can be done it allows a much wider choice of processes for acid gas removal, including conventional amine scrubbing as well as adsorption/oxidation type systems such as Stretford, Takahax, or IFP.

The adsorption/oxidation system could also be used for cleaning up the CO_2 vent stream, since it will remove H_2S without removing CO_2. Other methods may be cheaper or simpler, such as scrubbing with limestone slurry. This would pick up sulfur and have to be disposed of in a suitable manner. One possibility is to return it to the gasification zone. If flue gas desulfurization is used on furnaces in the plant, it is quite possible that the CO_2 vent stream could be included along with the flue gas for processing.

As described more fully in the section on Gas Handling and Sour Water Considerations, there appears to be a large potential advantage for developing practical techniques to remove sulfur at high temperature and avoid the need for liquid scrubbing. The major potential advantage would be to methanate CO directly, without having to shift or cool the gas for acid gas removal. It would also be necessary to remove dust at high temperature, but this could be done with sand bed filters. Thus the gas might first be cooled to about 600-900°F, and the dust removed. Then it would be desulfurized and

sent through the methanation reactor, where the CO would be reacted with steam and whatever hydrogen is present to form methane and CO_2. Finally the gas would be cooled and scrubbed for CO_2 removal and dried to provide pipeline quality gas. With this route the gas would never have to be reheated and cooled again, as is necessary with the present conventional systems.

Efficiency of the process and fuel consumption might be improved by reoptimizing a number of general items which are more or less conventional, such as the use of heat pumps, air-fin exchangers, and increased air preheat on furnaces (4,5).

12. TECHNOLOGY NEEDS

From this review and examination of environmental aspects of the BI-GAS process, a number of areas have been defined where further information is needed in order to evaluate the situation, or where additional studies or experimental work could lead to a significant improvement from the standpoint of environmental controls, energy consumption, or thermal efficiency of the process. Items of this nature will be discussed in this section of the report, and a summary is shown in Table 9.

Any coal conversion operation has solid refuse to be disposed of. Coal cleaning for the present design generates over 860 acre feet per year of refuse. In addition, the production of slag from gasification is 820 tons per day or another 10 acre ft/yr. More work is needed in order to define methods of disposal that do not create problems due to leaching of acids, organics, or sulfur which could contaminate natural water. In addition, adequate controls are needed with regard to the potential dust nuisance and washing away of particulates. In many cases the material may be suitable for land fill with revegetation. Although there is already a lot of background on this subject, specific information is needed on each coal and for each specific location in order to allow thorough planning to be sure that the disposal will be environmentally sound.

Coal drying is used on most coal conversion processes; consequently, considerable effort is warranted to optimize the operation from the standpoints of fuel consumption, dust recovery, and volume of vent gas to be handled. It will often be attractive to burn high sulfur coal rather than clean gas fuel, and to include facilities for cleaning up the vent gases.

The need for a simple, efficient means of feeding coal to the high pressure gasifier has been apparent and has received considerable study. For pressure levels of 400-500 psig, lock hoppers have been used satisfactorily, although they are expensive. For systems at 1,000 psig it may be attractive to pump a water slurry of the coal in order to pressurize it, particularly if it is possible to then evaporate the slurry at high pressure and thereby supply steam to the process.

One item that is critical in the BI-GAS process is the need for efficient removal of dust from gas at high temperature. In general, this is required in any coal gasification system where the gas is shifted before it is cooled and scrubbed. An important advantage is that particulates are kept out of the sour water stream, and consequently it is easier to clean up. Sand bed filters are promising for dust removal from hot gases, although they have not been fully demonstrated commercially.

In the area of acid gas removal, systems based on amine or hot carbonate are not completely satisfactory and leave room for improvement. Amine scrubbing is not effective on carbonyl sulfide, while contaminants such as cyanide interfere with regeneration of the scrubbing liquid. Hot carbonate systems do remove carbonyl sulfide, but it is often difficult to provide a highly concentrated stream of H_2S to send to the sulfur plant. In addition the CO_2 stream vented to the atmosphere may contain too much sulfur. Adsorption/oxidation systems are often not effective on carbonyl

Table 9

Technology Needs

- Environmentally sound disposal of large amounts of refuse from coal cleaning and washing, with regard to dust, leaching and sediment, trace elements, land use, etc.

- An optimized design for coal drying to use low excess air and give maximum allowable coal preheat, with good dust recovery.

- An improved system to feed coal into high pressure zones, for example using a piston feeder on water slurry. Slurry could be evaporated in heated fluid bed to make steam for gasifier and preheat the coal feed. Light hydrocarbon might be used instead of water, and condensed for reuse.

- A simpler and more efficient process for acid gas removal which would provide an H_2S stream of high concentration (e.g. 50 vol. %) to the sulfur plant, while giving a separate clean stream of CO_2 that can be vented to the air. Desirable features to include:

  - good sulfur clean up, to a few ppm
  - a clean CO_2 vent stream that does not require incineration
  - low utilities consumption
  - little or no chemical purges to dispose of

- An effective process to remove sulfur at high temperature could lead to improvements, such as reacting CO directly with steam to form methane.

- Ways to handle COS, CS_2, thiophene, etc, that are usually present and may not be removed by many acid gas removal processes. Hydrolysis to H_2S is probably one good approach.

- Sour water cleanup. Most of it may be used for quenching, but some purge will probably be needed to remove trace elements and perhaps ammonia and phenols. There is a great need for a practical system to evaporate sour water to make steam for use in the gasifier, and a fluid bed system appears promising.

- Information on trace elements and techniques for their disposal.

  - Extent of volatility for specific process and coal.
  - Where they appear in gas clean up system, and in what form. They may collect on the char or sand bed filter and build up by recycling. Others may appear on shift catalyst and in sour water or acid gas removal.
  - Many may be toxic and require separation and decontamination treatment before disposal.
  - Leaching may occur on the slag or on refuse from coal cleaning. Information is needed to define the potential problem and to devise environmentally sound disposal techniques.

sulfide and in any event do not remove CO_2 as required, and therefore additional processing is needed. The available systems for acid gas removal have very high utility requirements, causing a significant loss in thermal efficiency for conversion of coal to clean fuel products. In addition there is often a waste stream of chemical scrubbing medium which may be difficult and expensive to dispose of.

Desirable objectives for an acid gas removal process can be summarized as follows:(a) good clean up of all forms of sulfur to give a stream high in sulfur concentration for processing in a Claus sulfur plant, (b) effective CO_2 removal while producing a vent stream satisfactorily low in sulfur and pollutants, (c) low utility and energy consumption, (d) no waste streams that present a disposal problem.

The need for a process to remove sulfur at high temperature has been discussed fully in preceding sections of this report. Systems based on dolomite or iron appear promising; however, they may give less complete sulfur removal than conventional scrubbing systems, in which case a methanation catalyst that is tolerant of higher sulfur (e.g. 50 ppm) may have to be developed. If the sand bed filtering technique could be incorporated to remove particulates at the same time that sulfur is removed, such systems would be even more attractive. A further need is to destroy or remove undesirable contaminants such as carbonyl sulfide, cyanides, and possibly phenol and ammonia. This function might also be provided by a high temperature gas cleanup system.

The need for a simple,effective method to clean up sour water for reuse is another item that is common to most fossil fuel conversion operations. Sour water generally contains sulfur compounds, ammonia, H_2S, phenol, thiocyanates, cyanides, traces of oil, etc. These are generally present in too high a concentration to allow going directly to biological oxidation, but their concentration is often too low to make recovery attractive. Particulates, if present, further complicate the processing of sour water. Usual techniques for clean up include sour water stripping to remove H_2S and ammonia, and in addition, extraction may be required to remove phenols and similar compounds. Such operations are large consumers of utilities and have a large effect on overall thermal efficiency. In most cases the net amount of sour water produced is less than the amount of steam consumed by reaction in gasification plus shift conversion, which suggests a way to dispose of sour water. One approach is to use the sour water as quench on the hot gas leaving the gasifier, as is done in this BI-GAS design. However, it is not clear that compounds such as phenol and ammonia will actually be destroyed by recycling, so they may have to be separated and withdrawn as by-products.

An alternative approach is to vaporize the sour water to make steam which can be used in the gasifier. In this case,compounds such as phenol should be destroyed and reach equilibrium concentration in the circulating sour water. It may not be practical to vaporize sour water in conventional equipment such as exchangers, due to severe fouling and corrosion problems. Therefore, new techniques may be required,and one possibility would be to vaporize the sour water by injecting it into a hot bed of fluidized solids. The system could be very similar to that proposed for evaporating a water slurry of coal feed as discussed in connection with Figure 3. In fact, sour water may be used in some cases to form the slurry.

On trace elements, information is needed on the amount vaporized in the gasifier and what happens to them, where they separate out and in what form, so that techniques can be worked out for recovering or disposing of the materials. Again specific information is needed for each coal and for each coal conversion process since operating conditions differ. In many cases, the trace elements may tend to recycle within the system and build up in concentration. This offers an interesting opportunity to perhaps recover some of them as useful by-products. The toxic nature of many of the volatile elements should be given careful consideration from the standpoint of emissions to the environment, as well as protection of personnel during operation and maintenance of the plant. Carcinogenicity of coal tar and other compounds present in trace amounts or formed during start up or upsets should also be evaluated.

Protection of personnel, especially during maintenance operations should be given careful attention, which will require that additional information be obtained. Thus, toxic elements that vaporize in the gasifier may condense in equipment such as piping and exchangers where they could create hazards during cleaning operations. This may apply particularly to sand bed filters and to shift conversion reactors.

In this specific BI-GAS design, there is no sour process water effluent from the plant which might contain trace elements. Moreover the slag is drained and disposed of as a moist solid rather than a slurry. On this basis the question of cleaning up waste water effluent does not apply. However, in an actual application there will very likely be a water effluent, and detailed study of the facilities for clean up will be needed. In any event, the water make-up that is brought to the plant will contain dissolved solids including sodium and calcium salts. Calcium salts may be precipitated during the water treating operation to form a sludge which can be disposed of with the other waste solids, but the fate of the sodium salts in the make-up water calls for further study. These will leave with the blowdown from the cooling tower. If the concentration of dissolved solids is too high in this blowdown water to allow discharging it to the river, then some suitable method of disposal will have to be worked out. On one proposed commercial plant, this has been handled by using an evaporation pond where the water is evaporated to dryness. The salts accumulate and will ultimately have to be disposed of. If they cannot be used or sold then it would seem logical to dispose of them in the ocean.

13. PROCESS DETAILS

Additional details on the process are given in Tables 10 through 16.

Table 10

Steam Balance for BI-GAS Base Case

1265 psig steam lb/hr

Generated		Consumed	
Shift Outlet WHB	545,700	Gasifier	410,100
	---	Shift	65,400
	---	O_2 preheater	70,200
TOTAL	545,700		545,700

600 psig steam lb/hr

Generated		Consumed	
Auxiliary boiler	1,329,400	Power gener.	606,400
	---	Acid gas remov.	430,700
	---	to 400 psig steam	292,300
TOTAL	1,329,400		1,329,400

400 psig steam lb/hr

Generated		Consumed	
From 600 psig	292,300	Coal feeding	9,600
Shift & Meth. WHB	1,056,100	SNG compressor	192,900
	---	Glycol dry.	4,700
	---	O_2 plant	743,400
	---	O_2 compressor	397,800
TOTAL	1,348,400		1,348,400

50 psig steam lb/hr

Generated		Consumed	
Raw gas WHB	600,600	Acid gas removal	833,100
Claus plant	70,000	BFW deaerator	86,400
Turbo generator	298,900	Building heat	50,000
TOTAL	969,500		969,500

Table 11

Water Requirements for BI-GAS Base Case

Cooling Water Circulation	GPM
Coal preparation	0
Gasification	450
Quench & dust removal	0
Shift conversion	0
Acid Gas removal	114,000
Methanation & drying	1,930
Oxygen plant	117,600
Sulfur plant	1,100
Power generation	27,500
	262,580

Cooling Tower Makeup	GPM
Drift Loss	526
Evaporation	5,252
Blowdown (net)	1,200
	6,978

Table 12

Electric Power Consumption

	KW
Coal preparation	24,000
Gasification	3,500
Quench & dust removal	0*
Shift conversion	0*
Acid gas removal	0*
Methanation & drying	2,250
Oxygen plant	0*
Sulfur plant	270
Condensate pumps	4,700
Cooling Water pumps	7,140
	41,860

* A small amount of power will be used for instruments, lights, etc.

Table 13

Make Up Chemicals and Catalyst Requirements

Chemicals

Acid Gas Removal:

- scrubbing solution
- additives

Sulfur Plant tail gas cleanup

Glycol for drying prod. gas

Cooling Tower Additives

Anticorrosion, e.g. chromate
Antifouling e.g. chlorine

Water Treating

Lime
Alum
Caustic
Sulfuric acid

Catalysts, etc.

Sand for sand bed filters

Shift catalyst

ZnO guard bed to remove sulfur

Methanation catalyst

Molecular sieve to clean up CO_2 vent

Ion exchange resin for water treating

Table 14

Potential Odor Emissions

Coal storage and handling.
Coal preparation, washing, settling pond.
Coal drying - vent gas.
Vent gas from lock hoppers.
Wet ash handling and disposal.
Sour water stripping and handling.
CO_2 vent stream from acid gas removal.
Sulfur plant and tail gas.
Biox pond and other ponds.
Leaks: ammonia, H_2S, phenols, etc.

Table 15

Potential Noise Problems

Coal handling and conveyors.
Coal crushing, drying and grinding.
Oxygen plant air and oxygen compressors.
Lock hoppers, especially on depressuring from 1100 psig.
Burners on furnaces.
Stacks emitting flue gases.
Turbo-generator etc., in utilities area.

Table 16

Miscellaneous Inputs

For water treating: lime, caustic, alum, sulfuric acid, chlorine

Cooling water additives: anti algae (chlorine)
anti corrosion (chromium salt)

Other chemicals: carbonate and additives for acid gas removal
glycol for drying product gas

Catalysts, etc.: sand for sand bed filters
methanation catalyst
Claus plant catalyst
ZnO guard bed to remove sulfur
Sieve for sulfur clean up on CO_2 vent gas

Oil: to lubricate pumps, compressor, etc.

Biox nutrients, if required.

14. QUALIFICATIONS

As pointed out, this study does not consider cost or economics. Also, areas such as coal mining and general offsites are excluded, as well as miscellaneous small utility consumers such as instruments, lighting etc. These will be similar and common to all coal conversion operations.

The study is based on the specific process design and coal type cited, with modifications as discussed. Plant location is an important item of the basis and is not always specified in detail. It will affect items such as the air and water conditions available, and the type of pollution control needed. For example, this BI-GAS study uses high sulfur western Kentucky coal to supply gasification as well as utility furnaces. Therefore, flue gas clean up has been added. Because of variations in coal feed, moisture content, and other basic items, great caution is needed in making comparisons between coal gasification processes as they are not on a completely comparable basis.

Other gasification processes may make large amounts of various by-products such as tar, naphtha, phenols, and ammonia. The disposition and value of these must be taken into account relative, to the increased coal consumption that results and the corresponding improvement in overall thermal efficiency. Such variability further increases the difficulty of making meaningful comparisons between processes.

The BI-GAS process makes no appreciable amounts of tar, naphtha, or phenols; however, there could be a sizeable yield of ammonia, amounting to over 100 tpd and it is assumed that this can be recovered and sold.

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